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ASBESTOS-CEMENT PRODUCTS IN CONTACT WITH DRINKING WATER: SEM OBSERVATIONS

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Abstract

In studying the health effects of asbestos fibers ingested in drinking water it is important to know whether water can corrode the surface of asbestos-cement products to facilitate the release of the fibers to the water. Also, in the case of asbestos-cement pipe, it is important to know whether chemical inhibitors can coat the surface of the pipe to curtail this release.

Because of these needs, the effects of water on the surfaces of three asbestos-cement materials, namely, water pipes, cistern collecting tiles, and reservoir parapet walls, were studied using the Scanning Electron Microscope (SEM) and Energy Dispersive X-ray Analysis (EDXA). The micrographs from the SEM showed that the surfaces can be corroded, exposing and releasing fibers to the drinking water. The EDXA spectra showed that the surfaces of the asbestos cement materials can be chemically altered to cause the release of fibers. Also, these spectra showed that certain chemical inhibitors, under specific conditions, can coat the surfaces of pipe and presumably impede any fiber release.

KEY WORDS: Asbestos, Asbestos-Cement, Chrysotile, Specimen Preparation, Electron Microscopy, X-ray Microanalysis, Drinking Water, Water Supply, Water Pipe

Introduction

No doubt exists that exposure to inhaled asbestos fibers significantly increases the risk of lung cancer and pleural mesothelioma, especially among those who smoke. Higher than expected incidence rates of peritoneal mesothelioma, gastric, kidney, and colon cancer among workers occupationally exposed to airborne asbestos suggest that ingested asbestos may be a hazard because many inhaled asbestos particles are cleared from the respiratory tract and swallowed. G. W. Wright, the chairman of the American Water Works Association Research Foundation Committee (RFC), more recently reviewed the status of non-respiratory malignant asbestos-related disease and concluded that gastrointestinal cancer is dose related to the occupational exposure.[1] Also, chrysotile fibers up to 35 micrometers long were found in the kidney of a newborn baboon fed previously characterized reference asbestos suspended in formula.[2] Another study of the chronic effects of ingested asbestos in rats concluded that chrysotile asbestos could penetrate the gastrointestinal tract and migrate to the omentum, brain, lung, liver, blood, and kidney.[3] A 6-year San Francisco Bay area cancer study showed a statistically significant association between asbestos levels in the drinking water and cancers of the digestive tract.[4]

Because of these findings, the corrosion of asbestos-cement products and their release of asbestos fibers are important in the study of human health effects. Three asbestos cement products that come in contact with drinking water were studied to determine the effects of water on the surfaces.

The first samples studied were of asbestos cement pipes. Four field samples of A/C pipe and nine A/C pipe coupons used for in-house laboratory experiments were examined. Asbestos cement pipe is manufactured from a mixture of chrysotile asbestos with a small percentage of crocidolite, standard Portland cement, and silica sand. The concentration of asbestos employed in the mixture is from 15 to 20% and owing to its fibrous structure, it serves to bind and reinforce the cement during the forming and setting of the finished product.[5] All thirteen of the pipe sections were studied on the Scanning Electron Microscope (SEM) for structural changes and three

samples were studied by Energy Dispersive X-ray Analysis (EDXA) for changes in their chemical composition.

The second group of samples studied included a weathered and unweathered portion of an asbestos roofing tile which was used to collect water from a house for a cistern in southwest Ohio. The changes in its structure and chemical composition due to the weathering action of a period of over 15 years were observed.

Thirdly, a sample from a parapet wall was studied. Asbestos-cement building materials are occasionally used in water supply reservoir construction. Small reservoirs are sometimes covered with corrugated asbestos-cement sheets and some large reservoirs use the sheets as parapet walls around the reservoir to ward off animals. Both sides of a piece of asbestos cement sheet that had been used for some time as a part of a reservoir parapet wall were examined. This reservoir is the main source of the city's drinking water. The wall was within a foot of the water with the smooth side of the sheet facing outward. The side facing in toward the water was corrugated and covered with small raised pimples (approximately 2 mm in diameter), apparently a result of the form used to make the asbestos-cement sheet. Again the surfaces were examined for structural and chemical difference.

Sample Preparation

For analysis of chemical composition of the surface of the A/C samples, random pieces of each sample, approximately 12 mm by 6 mm, were mounted on 24 mm by 5 mm copper boats using a conductive graphite adhesive. These samples were then carbon coated in a Denton* vacuum evaporator. A sample for determining background counts was also prepared using only a copper boat and graphite adhesive. Each sample was then placed in a JEOL 100B Transmission Electron Microscope (TEM) operated at an accelerating voltage of 40 kV and a beam current of 55 - 60 μA.+ Using the SEM mode and a magnification of 300 X, the specimen was examined both longitudinally and laterally in a regularly-spaced rectangular array with seven to ten consecutive areas the size of 0.1 mm^2 being analyzed by the EDXA. Count rates ranged from 52 to 444 counts per second (CPS) for a time period of 500 seconds.

To examine the surface structure, random pieces from the same samples were coated with approximately 450 Å of gold-palladium in the vacuum evaporator. The samples were then placed in an ETEC (SEM) operated under an accelerating voltage of 20 kV, beam current of 150 μA,+ a working distance of 20 - 22 mm and at magnifications of 100 X, 300 X, 700 X, and 1,000 X. The samples were examined for the presence of asbestos fibers and degree of deterioration.

For the parapet wall and the roofing tile the percentage of exposed fibers at the surface with respect to the total area, or areal percentge of exposed fibers, was also determined. This was done by means of an adaptation of the areal method used in estimating mineral volume - percentages in petrology.[6] The edge of a photograph taken of the area under examination at 100X (0.5 mm^2) was traced on transparent plastic using a marker pen. The area covered by fibers or fiber bundles was then outlined. In the situation where only a few fibers were exposed, the photo was moved and the fibers were laid next to each other and outlined starting from one of the corners. When all the fibers were accounted for, the photo was removed and a sheet of squared paper was placed under the overlay. The number of squares covered by the fiber area was multiplied by 100 to give a percentage of the area covered by exposed fibers. An average value was calculated from three random photographs for the unweathered and weathered portions of the shingle and the smooth and corrugated sides of the parapet wall.

*The use of a specific manufacturer's name is for identification purposes only and does not constitute endorsement by the U.S. Environmental Protection Agency.

+Emission current near filament.

Results

The asbestos pipe samples analyzed showed a difference in both structural appearance and chemical composition. The micrographs of the unused pipe (#13) show that it was not a smooth surface (Figure 1), but rather a rough surface exhibiting some pitting. Also some asbestos fibers were exposed but these were few in number and the exposed portions of the fibers were under 10 micrometers in length and approximately 0.5 micrometers in width. The EDXA of the surface of the sample of unused pipe (Figure 2) shows a high concentration of calcium with small, but equal peaks of sulfur and silicon and a very small amount of iron. The count rates, from 6 separate analyses, showed calcium ranging from 27 to 33 times as high as iron and 7 to 9 times as high as sulfur and silicon. Also no magnesium was detected since few asbestos fibers, which contain magnesium, were exposed.

The American Water Works Association standard for A/C pressure pipe has an aggressiveness index (A.I.) for determining the suitability of using A/C pipe. The aggressiveness of water transported through a pipe, within the temperature range of 40-80°F, is determined by the formula: A.I. = pH + Log_{10}(AH).[7] The pH is given in standard pH units. The alkalinity (A) is total alkalinity in mg/l as calcium carbonate. The calcium hardness (H) is only the calcium constituent of the total hardness in mg/l as calcium carbonate. Higher values of this aggressiveness index are less corrosive than lower values. Water with an A.I. less than 10 is considered very aggressive to many types of pipe while A.I.'s greater than 12 are considered essentially non-aggressive.

The pipe sample (#12) from a laboratory experiment that was exposed to the aggressive water with an A.I. of 9.3 showed a structural change (Figure 3). There was more pitting and many more and larger fibers exposed at the surface. Fiber bundles up to 200 micrometers in length and 30 micrometers in width were seen. The EDXA (Figure 4) of this sample showed a marked decrease in the relationship of the calcium peak to other peaks as compared to the unused pipe. There also was an

increase of both silicon and iron; the peaks of both were about twice as large as the calcium. Also magnesium was detected as would be expected with more fibers exposed. Small amounts of aluminum, manganese and titanium were also found. Some trace elements may have come from the corrosion of the stainless steel tanks used in the experiment.

The pipe sample (#4) exposed to water that had zinc chloride added as an inhibitor showed a very smooth surface as compared to the other samples (Figure 5). The surface was not totally smooth, but the reason may have been the fact that the pipe wasn't exposed to the zinc long enough to be totally coated. This experiment ran for six months. Very few fibers were observed. The EDXA (Figure 6) showed a high amount of calcium, almost 3 times as high as the silicon and iron peaks. Trace amounts of magnesium and lead were found and a very small chlorine peak was observed. The spectra show a large zinc peak indicating the zinc is the primary coating material. In a previously published study[5] of additives to prevent corrosion of asbestos-cement pipe, zinc orthophosphate was added to a pipe test system. EDXA analysis showed zinc coating the surface of the pipe, but the anion, in this case, phosphate, could not be detected.

Field samples of A/C pipe were also studied to investigate situations in which the release of fibers to the drinking water is inhibited (Table 1). In one Massachusetts town, the water was soft and had a very low aggressiveness index (A.I. = 7.46). An A.I. under 10 is generally thought to be very corrosive to A/C pipe. However, no significant numbers of asbestos fibers were found in consumers' tap water which had flowed through A/C pipe. SEM-EDXA analysis showed very few exposed fibers and an overall coating of iron on the pipe's surface. In a similar situation in Connecticut, no significant numbers of fibers were found after mildly aggressive water (A.I. = 11.1) had flowed through A/C pipe. Again the SEM-EDXA analysis showed no exposed fibers on the pipe's surface, but the coating consisted of manganese rather than iron. In a South Carolina town the addition of zinc orthophosphate to prevent fiber release from A/C pipe was field evaluated. Two sections of A/C pipe, an eight-inch diameter and a six-inch diameter, were analyzed. The SEM-EDXA analysis showed that the pipe section to which the highest concentration of zinc (0.3 mg/L) had been added had the best coating.

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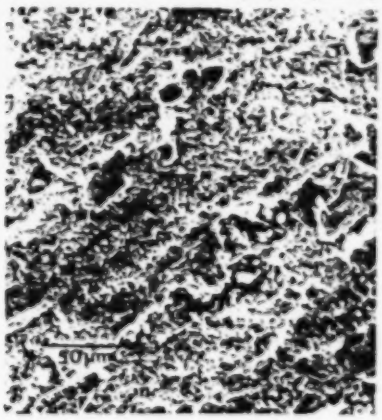


Fig. 1. SEM of the surface of unused A/C pipe

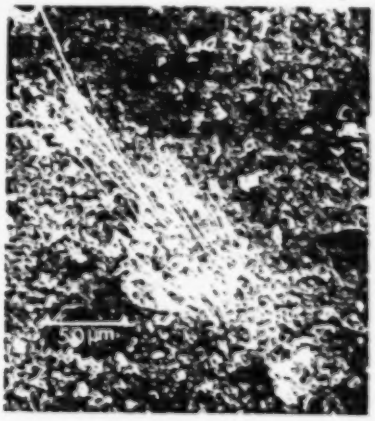


Fig. 3. SEM of the surface of A/C pipe #12, exposed to aggressive water

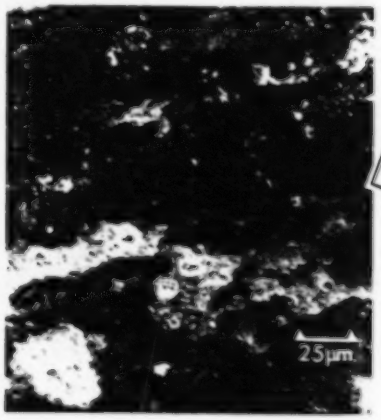


Fig. 5. SEM of the surface of A/C pipe #4, exposed to water with zinc chloride added as inhibitor

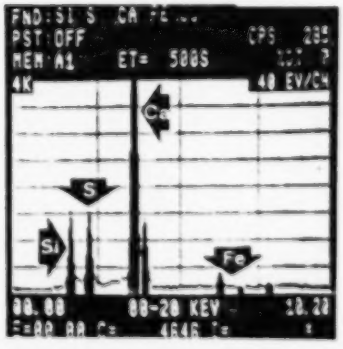


Fig. 2. Energy Dispersive X-ray spectrum of pipe surface as in Fig. 1.

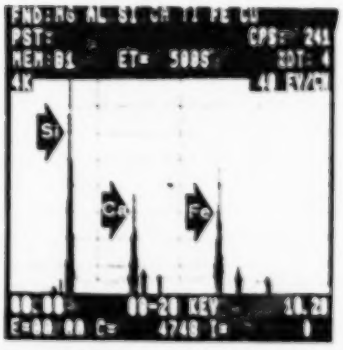


Fig. 4. Energy Dispersive X-ray spectrum of pipe surface as in Fig. 3.

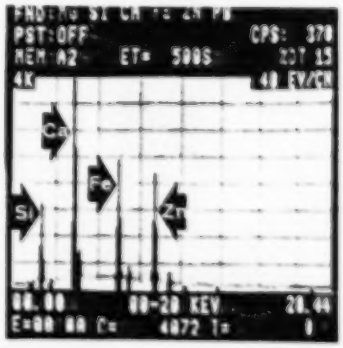


Fig. 6. Energy Dispersive X-ray analyses of pipe surface as in Fig. 5.

Table I. Field Studies A/C Pipe

Sample	SEM Observation
City in Massachusetts A.I. = 7.46 No inhibitors added at plant No fibers found in drinking water	Very few exposed fibers Coated by iron
City in Connecticut A.I. = 11.1 No inhibitors added at plant No fibers found in drinking water	No exposed fibers Coated by Mn
City in South Carolina A.I. = 10.4 Zinc orthophosphate added 0.1 mg/L zinc Fibers found in drinking water before treatment	A few fiber bundles Some pitting
City in South Carolina A.I. - 10.4 0.3 mg/L zinc Fibers found in drinking water before treatment	Very few exposed fibers Very little deterioration

In all, nine pipe coupons from laboratory experiments were analyzed by SEM. The SEM micrographs on pipes (#12 and #10) showed that water can corrode the surface to expose fibers. Three pipes (#8, #20, #22) all had Virchem® (zinc orthophosphate) added, but the micrographs showed that the zinc coats better under certain conditions of pH and A.I. than others. The same results were observed on the SEM micrographs of the pipes (#1, #9, #4) where zinc chloride was added. Again different water conditions seemed to facilitate the coating of zinc onto the pipe (Table II).

As a corollary to the pipe samples observed by SEM-EDXA analyses, two modes of EDXA analysis were run on unused pipe to compare results. The first was the spot mode, covering an area of 200 Å at 300X. Eight analyses were run on random points of the sample with five of them being consistent, showing a large amount of calcium with small amounts of silicon and sulfur. Two of the analyses showed silicon as high as calcium and one analysis showed practically no silicon or sulfur. Seven analyses in the scan mode, covering an area of 0.1 mm^2 at 300X, showed consistent spectra with high calcium and smaller but equal silicon and sulfur.

The roofing tile and parapet wall were analyzed by the SEM for surface structural change and the results are summarized in Table III. A significant difference in percentages of exposed fibers was observed between the unweathered (Figure 7) and weathered (Figure 8) portion of the roofing tile as well as the smooth (Figure 9) and corrugated (Figure 10) side of the parapet wall.

The weathered roofing tile showed, on average, 27% exposed fibers as compared to only 2% exposed fibers in the unweathered tile. The corrugated side of the parapet wall showed 36% exposed fibers as compared to less than 1% exposed fibers on the smooth portion of the wall. The EDXA analysis of all four samples were relatively the same, with iron, silicon and calcium being the major peaks. The only difference was a slight decrease in calcium on the weathered portion of the roofing tile.

Table II. Inhouse A/C Pipe Coupon Studies

Number and Sample	A.I.	SEM Observations
13 Unused pipe (Fig. 1)		Rough surface A few fibers exposed
12 No corrosion control (Fig.3)	9.3	Some deterioration Fiber bundles exposed
10 No corrosion control	10.2	Some deterioration Fiber bundles exposed
8 Virchem ®* added	9.3	Some deterioration A few fiber bundles
20 Virchem®* added	10.2	Surface coated very few fibers
22 Virchem®* added Zn 1.0mg/l for 9 days	10.5	Surface coated very few fibers
4 Zinc chloride added (Fig.5)	9.8	Some deterioration Some fiber bundles
9 Zinc chloride added, Zn 0.7mg/l first, always 0.3mg/l		Very good coating Smooth surface Very few fibers

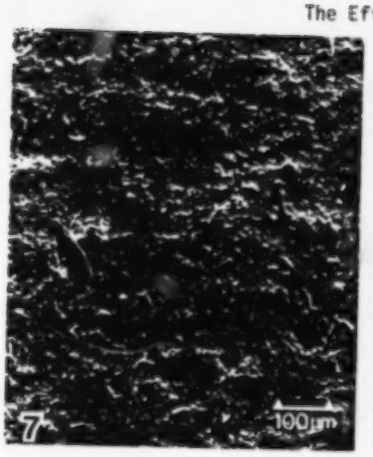
*Virchem ® = Zinc orthophosphate

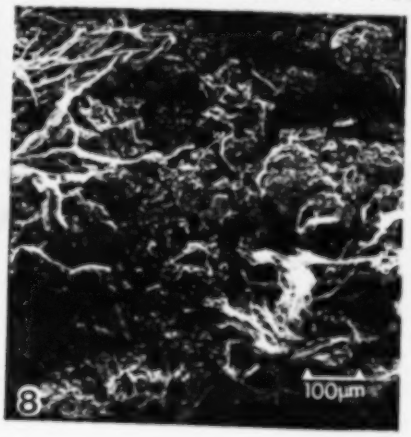
Table III. Percentage of Surface Area Covered by Exposed Fibers

	1	2	3	Ave.	Fig.No.
Cistern tile: weathered	28%	26%	26%	27%	8
Cistern Tile: unweathered	2%	2%	2%	2%	7
Parapet wall: toward reservoir	46%	26%	-	36%	10
Parapet wall:	0.3%	0.4%	-	1%	9

Conclusions

Water can corrode the surface of asbestos cement materials to release asbestos into the drinking water. The SEM micrographs of the weathered portion of the roofing tile showed many exposed fibers. Analysis by TEM of water collected directly from the roof during a rain showed a concentration of 420 million fibers of chrysotile asbestos per liter. Drinking water, which was collected in the home's cistern, showed a concentration of 96 million fibers of chrysotile per liter. In a case such as this it would be prudent to remove the asbestos tile or apply a sealant to coat the remaining fibers.





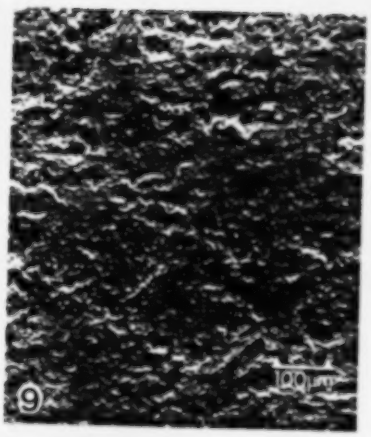
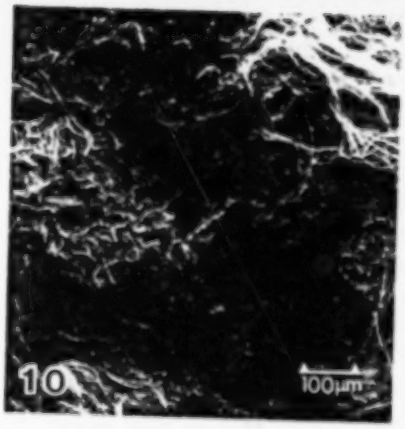


Fig. 7. SEM of the surface of unweathered roofing tile.

Fig. 8. SEM of the surface of weathered roofing tile.

Fig. 9. SEM of surface of the smooth side of parapet wall.

Fig. 10. SEM of the surface of the corrugated side of the parapet wall toward the reservoir.



In the case of the parapet wall, the SEM micrographs showed fibers on the surface facing the reservoir water. Samples taken from the reservoir at different times had widely varying asbestos fiber concentrations when analyzed by TEM. The variance in the fiber concentrations may be a result of intermittent weathering and erosion of fibers from the parapet wall. Again, it would be prudent to coat the surface with a sealant to prevent the release of fibers.

The SEM-EDXA analyses of the A/C pipe coupons show that water can corrode the surface of the pipe to expose the asbestos fibers. In some cases it may be necessary to coat the pipe with inhibitors. Various other inhibitors added under different water conditions should be analyzed by the SEM and EDXA as to their effectiveness.

To obtain the most consistent results for the chemical composition of the pipe surface by EDXA analysis it is better to use the full scan at low magnification rather than the spot mode which may give varying results.

The SEM-EDXA analyses show very explicitly the effects of water on the surface of asbestos-cement materials. Even though it has been said that asbestos-cement materials reach an equilibrium after a few years of contact with water, the micrographs and spectra show a deterioration after 15 years exposure, as in the case of the roofing tile. Further studies will have to be done to help determine more explicitly the cause of the deterioration and the possible solutions to inhibit the release of asbestos fibers.

Authors' late addition: A relevant recent paper is "The behavior of asbestos-cement pipe under various water quality conditions: a progress report" by R.W. Buelow, J.R. Millette, et al., J. Amer. Water Works Assoc., 5:72(2)91-102 (1980). This gives the details of the experimental setup for the asbestos-cement pipe coupon tests.

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Discussion with Reviewers

E. J. Chatfield: The reference you quote for the association between asbestos levels in drinking water and gastrointestinal cancer is one which finds a positive association. Other studies exist (Wigle, D.T., Cancer Mortality in relation to asbestos in municipal water supplies. Arch. Environ. Health, (1977) 185-190) in which negative conclusions are drawn. Are you aware of any other studies which gave either positive or negative results for this association?

Authors: A few epidemiology studies of persons exposed to asbestos in their drinking water have been published. Two studies have examined the population of Duluth, Minnesota, and concluded that no carcinogenic effect was apparent in the first 15 years following the dumping of amphibole fiber contaminated mine tailings into Lake Superior, but the authors of both studies cautioned that the period of observation was short compared to the latent period for occupationally induced asbestos carcinogenesis (20-30 yrs.). (Mason, T.J., et al., Asbestos-like fibers in Duluth Water Supply. Relation to Cancer Mortality. J. Amer. Med. Assoc. 228:1019-1020 (1974) and Levy, B.S., et al., Investigating Possible Effects of Asbestos in City Water: Surveillance of Gastrointestinal Cancer Incidence in Duluth, MN. Amer. J. Epi. 103:362-368 (1976)). Another study failed to show any relationship between cancer and the use of asbestos-cement pipe in Connecticut (Meigs, J.W., et al., Asbestos Cement Pipe and Cancer in Connecticut, 1955-1974. J. Envir. Health 42(4):187-191 (1980)). Asbestos analyses in each Connecticut town with A/C pipe showed low concentrations of fibers in the water. It is interesting to note that while Wigle (1977) concluded that there was no definitive evidence of an association between cancer and asbestos in water supplies in Quebec, he did find excess mortality due to cancer of the stomach (men), pancreas (women), and lung (men) in the two communities with known high exposure. The California study (Reference 4) found statistically significant positive trends of increased risk for cancer of the stomach (male), peritoneal (female), lung (male), gall bladder (female) and esophageal (female). Case-control studies, a more definitive epidemiology tool, are either underway or planned for two areas with high asbestos in the drinking water: around San Francisco, CA, USA and around Seattle, WA, USA.

E. J. Chatfield: Would it be true to say that perhaps the A.I. may not be a completely adequate index of the corrosive potential of water on A/C products? Do your results indicate any other variables which should be included in this index?

Authors: The A.I. is not a completely adequate index of the corrosive potential of water on A/C products. Other variables such as the iron and zinc concentrations are also important.

E. J. Chatfield: What was the purpose of the sample consisting of only a copper boat and graphite adhesive?

Authors: The copper boat with graphite adhesive was analyzed by itself to see if any elements were detected which would show up on the elemental analysis of the A/C pipe. Only the copper from the boat was detected.

E. S. Boatman: Your surface density estimates of exposed fibers in samples of cistern tile and parapet wall were determined at a photographic magnification of 500 X. What was the smallest fiber size you could observe under these conditions? In view of the limited number of fields observed (2 to 3 photographs), the results of percentages of surface area covered by exposed fibers are remarkably close. One would expect a greater variability. Would you comment on these two points?

Authors: Areal percentage was determined at a photographic magnification of 100 X with the smallest fiber observed being 1.0 μm in width. This method was used primarily to show the great difference in exposed asbestos fibers for the different samples. When any of the samples were examined at 1000 X, fibers appeared in bundles rather than individual fibrils sticking out of the matrix. Even if all the possible small fibers were counted, the changes in areal percentage would not have been great and in no way would have changed the large difference between the two samples in each group. After scanning the total specimen area, the pictures taken and used were those believed to be representative of the whole sample. Had variability been great, several more areas would have been photographed for determination of areal percentages to yield a representative average value.

E. S. Boatman: In view of the observation that the smooth surface of the parapet wall showed so few exposed asbestos fibers would you say that the action of weathering is less important as to the release of fibers than surface imperfections arising during manufacture of the sheets?

Authors: The surface imperfections in a manufactured asbestos-cement sheet are very important to the release of fibers and would tend to accelerate the action of weathering to corrode the cement sheet.

K. S. Chopra: How would you check the effect of water in real world samples such as wall, pipe, etc.? Could you propose an optical method also?

Authors: TEM analyses for asbestos fibers from water samples that had flowed through A/C pipe can show if the water is corrosive. Also, there is a physical scratch test as implied in Reference 7. Optical methods could be developed for very corroded pipe with large fiber bundles exposed, but for research on coating efficacy other methods have to be used.

K. S. Chopra: What is the size of field in case of EDXA spectrums shown in Figures 2, 4 and 6?

Authors: An area the size of 0.1 mm^2 was analyzed for the EDXA spectra.